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THE MECHANISM OF ADHESION BETWEEN
SOLID AEROSOL PARTICLES

by

Dr. T. G. Owe Berg

Investigations Under

U. S. Army Chemical Center

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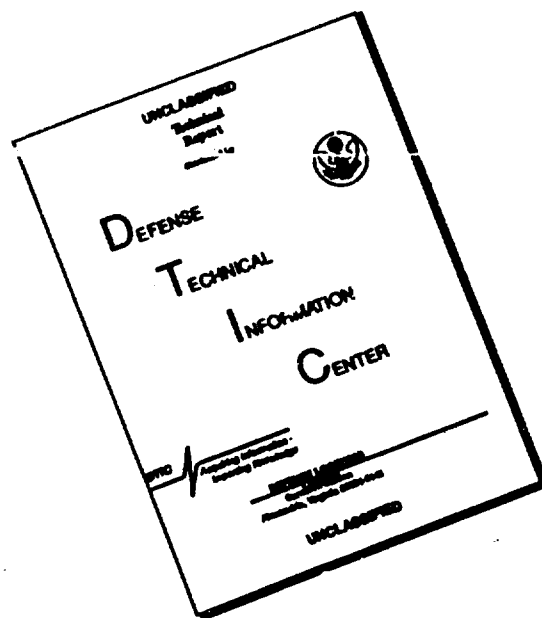
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FOREWORD

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THE MECHANISM OF ADHESION BETWEEN SOLID AEROSOL PARTICLES

by

T. G. Owe Berg

Abstract

The results of a review of the literature and of a few supplementary experiments on the adhesion between solid bodies and between solid particles are reported. Water is the cause of adhesion under ordinary conditions, namely, by three mechanisms:

- a. Capillary forces (surface tension) in the case of comparatively thick films of water on insoluble solids.
- b. Intermolecular forces (hydrogen bonds) between water molecules and OH groups in the solids in the case of comparatively thin (monomolecular) films on insoluble solids.
- c. Coalescence by dissolution and precipitation in the cases of thin or thick films of water on soluble solids.

The role of electrostatic charge is not well clarified by the experimental evidence. It appears that electrostatic attraction after contact contributes negligibly, and that the main effect of electrostatic charge is that of a discharge current upon the mechanism c.

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1. INTRODUCTION

In preparing an experimental study of the agglomeration of solid aerosol particles, a review has been made of the literature on agglomeration of solid particles and the adhesion between solid bodies. The review is based upon the index of the Chemical Abstracts back to 1906, Volume 1, and the references given in the papers listed. This report presents the evidence collected and conclusions drawn from this evidence. References are given to papers deemed particularly informative. Completeness of references has not been considered essential to the purpose. In addition, a few exploratory experiments have been conducted to check and to supplement the information obtained from the literature.

The subject has been treated in the literature with a view to a variety of applications in addition to aerosols, e.g., grinding of powders, flow of powders, caking of fertilizers, flotation of ores, formation of hail, sintering and solid-state reactions, friction, but also from more general considerations and in connection with various theories. These treatments are sometimes based upon premises that are inapplicable to aerosols, and their bearing upon the agglomeration of solid aerosol particles is then limited to the most basic elements.

For the purpose at hand, it is convenient to distinguish between freshly formed surfaces and aged surfaces of solid particles and solid bodies. To this distinction corresponds one between phenomena in the formation of particulates and properties of airborne particulates. The formation of particulates will not be treated in this report. This distinction is a logical consequence of the peculiar physical and chemical properties of freshly formed surfaces as compared to aged surfaces and of the profound effects of these properties upon agglomeration. Accordingly, information on agglomeration in grinding and in friction will not be considered further than incidentally in this report.

A very special case of agglomeration occurs in wash-out, when aerosol particles are captured by liquid (e.g., rain) drops and held in solution or suspension during the agglomeration process. In this case, the aerosol is virtually converted into a hydrosol, a subject that does not fall within the scope of this report.

Adhesion between two particles requires that they be brought in contact with each other, either by random collision or by long-range forces, and that they be kept in contact with each other by short-range forces. The establishment of contact will not be considered in this report. The attention will instead be focused upon the phenomena after establishment of contact and especially upon the short-range forces operative on solid bodies in contact with each other.

The experimental evidence on the agglomeration of powders and the adhesion between solid bodies shows unambiguously that water is instrumental in both cases. Occasionally, water may be replaced by some other liquid. Essential is the presence of a thin film of liquid at the contact. The evidence indicates several mechanisms for this effect of water. A comparatively thick film of water appears to act by its surface tension, i. e., by capillary forces. A monomolecular film of water appears to act by the formation of hydrogen bonds between OH groups in the hydrated surfaces. Finally, the bodies in contact may dissolve in the water and recrystallize out of the water so that a bridge is formed across the interface between the two bodies that eventually leads to their coalescence. These various cases will be discussed in Sections 2 to 5.

The experimental evidence on the role of electrostatic charges and forces in adhesion and agglomeration is inconclusive. It is not even clear whether short-range electrostatic forces exist under ordinary experimental conditions. This subject will be discussed in Section 6.

2. ADHESION BETWEEN LARGE SOLID BODIES

For the purpose of this report, experiments on the adhesion between large solid bodies, in the millimeter or centimeter range, are informative as model experiments. A number of such experiments have been performed explicitly to this purpose. Among those there are a few of particular interest.

A well-known precision measuring tool consists of a series of accurately machined steel blocks, several of which can be wrung together so that they form one unit without interspace and holding together so strongly, that they cannot be pulled apart but can only be separated by sliding them off the composite block. These are stored with greased or oiled surfaces for protection. When used, this grease or oil is, of course, removed, but if they are dried too carefully, they do not adhere one to the other. This adhesion was studied by Budgett¹⁾, who found that adhesion requires a thin film of liquid. Budgett measured the force of adhesion with water and various organic liquids. The force was strongest with water. There was no difference between the forces measured in the ambient and in a vacuum. This rules out an effect of a vacuum between the surfaces. Surface tension of the liquid was estimated to account for no more than 4% of the force.

Stone²⁾ measured the adhesion between pairs of small glass beads, about 1 mm in diameter. One of the beads was suspended as a pendulum; the other bead was mounted on a horizontal micrometer screw. The two beads were brought into contact and then pulled by the micrometer screw until the weight of the deflected pendulum broke the adhesion. Adhesion occurred only when moisture was present. Thus, beads exposed to a current of dry air showed no adhesion but showed normal adhesion when breathed on. Beads adhering to each other and given a deflection of the pendulum corresponding to 75% of the force of adhesion could not be separated by exposure to dry air. When heated carefully in a flame, they softened and coalesced without breaking apart. There was no effect of electrostatic charge upon the force of adhesion.

Nakaya and Matsumoto³⁾ measured the force of adhesion between ice spheres using essentially the same technique as Stone. They found evidence to the presence of a film of water on the contact surfaces of the adhering surfaces. The adhering spheres slid readily on each other. An interesting observation, that will be further discussed in Section 4, is that a rotation occurred sometimes before separation. The angle of rotation varied, apparently in a random manner, between 6° and 47° . Hosler, Jensen, and Goldshlak⁴⁾ reported similar measurements in which the humidity of the surrounding atmosphere was varied. The force was much smaller in a dried atmosphere than in one saturated with respect to ice except, of course, in the vicinity of the melting point.

The experiments just referred to are too simple and straightforward for misinterpretation of the effect of the liquid. In a somewhat more complex experiment, reported by Cremer and her associates⁵⁾, the adhesion between powder particles in the range 2 to 300μ was determined from the sliding angle. This experiment will be discussed further in Section 3. It was found that the force of adhesion in MgCO_3 powder was reduced by a factor of 5 after drying at 110°C .

McFarlane and Tabor⁶⁾ confirmed the results of Budgett and Stone as to the effect of water. They used a bead suspended as a pendulum and a vertical flat plate. There was no adhesion with glass, Pt, or Ag in dry air, but there was strong adhesion in humid air, particularly with glass. A plot of the force against the relative humidity showed no force below 80% humidity and a rapid rise between 80% and 90% to a constant value for 90% to 100%. This force was the same as that measured when a small drop of water was introduced between the two surfaces. This indicates a comparatively thick liquid film and not a monomolecular film of adsorbed water.

Accordingly, the force was determined by surface tension. Using water and other liquids of surface tensions between 22 and 73, excellent agreement was found between the surface tension calculated from the measured force and that determined by other means. It is interesting to note that there was no adhesion in an atmosphere saturated with vapors of benzene or alcohol. In the case of a steel ball and an indium surface with lauric acid between the two surfaces, evidence was found to the effect that separation occurred by stretching and rupturing of a fatty acid monolayer. In this particular case, the adhesion was thus not attributable to surface tension.

It should be emphasized that the results of McFarlane and Tabor are not necessarily in disagreement with those of Budgett since the experimental techniques were different in the two cases: Budgett used flat surfaces that were wrung together so that a very thin film of liquid was formed; McFarlane and Tabor used a sphere and a flat that were gently approached one to the other so that a comparatively thick layer of liquid should have been left between the two. Reference may be given to Rolt and Barrell⁷⁾ as to the behavior of the liquid film between flats wrung together. With respect to the thickness of the water film, the experimental conditions of McFarlane and Tabor are more comparable to those of Stone.

An important observation by McFarlane and Tabor is that the force of adhesion between a steel ball and a metal plate was proportional to the contact surface when the two were pressed hard together. This case is similar to that studied by Budgett. In the case of surface tension in a thick film of liquid, the force is proportional to the radius of the ball.

Experiments similar to those of Stone have been reported by Tomlinson⁸⁾, Bradley⁹⁾, and Harper¹⁰⁾, who seem to have been unaware of the results of Stone and Budgett. Tomlinson and Bradley found, as did McFarlane and Tabor, that the force of adhesion between glass spheres of equal size is proportional to the radius. The constants of proportionality were 1600, 20, and 1000, respectively, when the force is expressed in dynes and the radius in centimeters. The data of Tomlinson and of McFarlane and Tabor are thus in fair agreement. This indicates a thick film of water in Tomlinson's experiment as well as in that of McFarlane and Tabor. Bradley's force constant differs by orders of magnitude. (Bradley used quartz spheres, not glass, but it is hard to believe that this accounts for the difference.) Bradley found twice this force constant for sodium borate spheres. He showed that the adhesion in that case was caused by water.

Bradley used spheres of different radii. He plotted the force against $\frac{d_1 d_2}{d_1 + d_2}$, where d_1 and d_2 are the diameters of the spheres. The plot is given as linear, but the spread of the data and the range covered by the data may well permit a power of $\frac{d_1 d_2}{d_1 + d_2}$ different from the first.

Thus, the area of contact between two elastic spheres pressed together by a force P is proportional to $P \left(\frac{d_1 d_2}{d_1 + d_2} \right)^{2/3}$. As already mentioned, McFarlane and Tabor found the force to be proportional to the contact area when a metal sphere was pressed hard against a metal flat. A simple explanation of the small force measured by Bradley is that he used an atmosphere of comparatively low humidity. As already mentioned, McFarlane and Tabor found no measurable force in a dry atmosphere.

Several investigators have measured the force of adhesion between glass flats and claimed to have found a distance force at a separation of about 1μ between the flats. Overbeek et al ¹¹⁾ give references to this work. There are drastic discrepancies between the results of the different investigators and also between the results of the same investigators ^{11, 12)}. Kitchener and Prosser ¹³⁾ measured forces of the order of 0.4 to 6 dynes/cm² at separations of 1.2 to 0.7 μ . They made a comment to the effect that a force of the order of dynes was reduced to 10^{-2} dynes after exposure to a discharge in less than 1 mm of air for 10 sec, and that pumping down to 10^{-5} mm reduced this force by another factor of 7 over a 24 hr period. After this treatment the force remained constant over a period of 72 hr. It was then assumed, somewhat arbitrarily, it appears, that in this state London - van der Waals forces alone were operative. The effect of reducing the pressure further, e. g., to 10^{-6} mm, was not reported. An interesting observation is that repulsion occurred at a separation smaller than 0.6 μ so that the attraction could be measured at greater separations only.

The purpose of the discharge in this experiment was to remove electrostatic charges. Overbeek et al ¹¹⁾ exposed their surfaces to water vapor to the same purpose.

Stone, Tomlinson, Bradley, and McFarlane and Tabor made sure that electrostatic forces did not contribute to their adhesion forces. Harper ¹⁰⁾ did not measure the force of adhesion but observed occasional sticking when two quartz spheres were brought in contact with each other and then separated. This process occasionally produced charge on the spheres, but there was no correlation between charging and sticking.

It seems to have been convincingly shown that the adhesion between solid bodies, particularly glass, is caused by water. It appears that this adhesion is caused by capillary forces (surface tension) in the case of thick films of water on the surfaces and by molecular forces, presumably hydrogen bonds between the water and OH groups in the surfaces ¹⁴), in the case of thin films of water.

Attempts at showing London - van der Waals forces, i. e., long-range attraction between the molecules in the two bodies, have failed. If such forces occur at all, they must be of a negligible order of magnitude as compared to other forces of adhesion ¹⁵).

Reference should finally be given to a paper by Howe, Benton, and Puddington ¹⁶) that unfortunately is too brief to convey a clear understanding of the experiment and the result. These investigators found a gradual buildup of adhesion between a Pyrex bead and a Pyrex plate over a period of several hours of contact and also a rapid decay of the adhesion over a period of several seconds. The experiment was conducted in vacuo or in N₂ of low pressures.

3. ADHESION BETWEEN POWDER PARTICLES

Cremer et al ⁵) have developed a technique for the measurement of adhesion and friction between powder particles that permits determination of the two quantities separately. This technique has been used in preliminary experiments in the author's laboratory. This section will be devoted to this technique and the results obtained. Measurements of the combined effects of adhesion and friction will be treated in Section 4.

A pile of powder is placed on a tiltable plane, the angle of inclination of which is increased until the powder slides off. The force of adhesion H is then

$$H = w \sin \alpha - \mu w \cos \alpha \quad (1)$$

when w is the weight of the powder, μ is the coefficient of friction, and α is the inclination. Varying w , Cremer plots $w \sin \alpha$ against $w \cos \alpha$. The plot is linear with a slope equal to μ and an intercept equal to H .

This formula is valid for a solid body on an inclined plane. When there is no force H , α is the friction angle, $\alpha = \arctan \mu$. If the pile of powder is taken as a solid body, H is the adhesion between the powder and the plane, and μ is the coefficient of friction between the pile of powder and the plane.

An experiment reported by Cremer with magnesite powder of particle size 80 to 300 μ on surfaces of glass, magnesite, Ni, and Mo shows that the product of force H and particle size d is a constant that is independent of the material of the plane. The values of μ are not given.

The fact that the product Hd is independent of the material in the plane indicates that H is not the force of adhesion between powder and plane but that between powder particles. In order to check this conclusion, a few simple experiments were conducted in our laboratory with silica sand on glass (microscope slide). Sieve fractions between 44 and 200 μ were used. The slide was clamped to a bevel protractor and raised until part of the sand slid off. The amount of sand put on the slide and the distribution of the sand over the slide surface were not controlled; the sand was merely scooped onto the slide.

When the sand was applied in a single layer, it slid off as individual grains and at an angle that was independent of the particle size. When the sand was applied in a thick layer or a pile, the bottom layer stuck more strongly to the slide, presumably as a result of the load on it, and the second and higher layers slid off this bottom layer. This occurred at a greater angle and at an angle that increased with decreasing particle size. When a large amount of sand was applied, the angle was independent of the amount applied. Hence, H is proportional to w. A plot of $\frac{1}{d}$ against α was then made and extrapolated to $\frac{1}{d} = 0$, i. e., infinite grain size. The plot was not far from linear, and the extrapolation was easily made. The extrapolation gives the angle of friction α_0 so that $\mu = \tan \alpha_0$. This value of μ was taken as a first approximation, and $\sin \alpha - \mu \cos \alpha$ was computed. A new plot was made of $\frac{1}{d}$ against $\sin \alpha - \mu \cos \alpha$. This plot did not extrapolate quite to the origin, but after a slight adjustment of μ a linear plot through the origin was obtained. The data in Table 1 show that the product $d(\sin \alpha - \mu \cos \alpha)$ is fairly constant.

Table 1. Adhesion in Silica Sand. $\mu = 0.48$

Particle Size (microns)	d (mm)	α	$d(\sin \alpha - \mu \cos \alpha)$	Deviation from average
175 - 208	0.191	28.3°	0.0086	- 0.0017
124 - 175	0.149	28.5°	0.0115	+ 0.0012
104 - 124	0.114	31.3°	0.0109	+ 0.0006
74 - 104	0.089	32.3°	0.0110	+ 0.0007
62 - 74	0.068	34.4°	0.0097	- 0.0006
44 - 62	0.053	40.7°	0.0103	0.0000

average 0.0103

The deviations from the average seem to be random, although they are largest for the largest particles.

It follows that

$$H = \text{const} \times \frac{w}{d} = \text{const} \times \text{particle surface area} \quad (2)$$

for if

$$H = k_1 w \quad (3)$$

and

$$\frac{1}{d} = k_2 (\sin \alpha - \mu \cos \alpha) \quad (4)$$

then, from Equation (1),

$$k_1 = \frac{1}{k_2 d} \quad (5)$$

In Cremer's plot of $w \sin \alpha$ against $w \cos \alpha$ for a given value of d , the intercept is clearly proportional to w . But the intercept is also proportional to $\frac{1}{d}$. Hence, H is proportional to $\frac{w}{d}$, i. e., to the particle surface area.

Our experiment was performed as a quick check of Cremer's experiment and essentially in order to observe how the powder slides. It confirms Cremer's results, which are superior in accuracy.

It seems thus to be established that the force of adhesion measured by this technique is that between powder particles, and that this force is proportional to the surface area of the powder particles.

An interesting observation made by Cremer is that with MgCO_3 the force of adhesion was reduced by a factor of 5 by drying at 110°C . This shows that water is instrumental in adhesion in this case as well as in the cases discussed in Section 2. A quick check on this point was made with the silica sand. The sand used in the experiments just described was placed in a dessicator for 15 days, whereupon the experiment was repeated. The results, shown in Table 2, were about the same as in the preceding experiment. The force constant k_2 was found to be 110 as compared to 95 before, i. e., the force of adhesion was reduced by 15%. The value of μ was unchanged, $\mu = 0.48$. The effect of drying was thus comparatively small in this experiment, presumably as a result of rapid readsorption of water on exposure to the ambient.

Table 2. Adhesion in Silica Sand. $\mu = 0.48$

Particle Size (microns)	d (mm)	β	α	d (sin α - μ cos α)	Deviation from Average
175 - 208	0.191	36°	28.0°	0.0087	- 0.0002
124 - 175	0.149	37°	29.0°	0.0095	+ 0.0006
104 - 124	0.114	38°	30.0°	0.0096	+ 0.0007
74 - 104	0.089	41°	31.0°	0.0092	+ 0.0003
62 - 74	0.068	37°	31.0°	0.0070	- 0.0019
44 - 62	0.053	39°	34.75°	0.0093	+ 0.0004

avg 0.0089

Cremer's technique has been used recently by Patat and Schmid¹⁷⁾ with astonishingly poor results. The spread of the data was such that correlations had to be looked for by means of "modern statistics". This appears to be explained by a drastic modification of the technique. The powder was distributed as uniformly as possible on the plane. This may mean that it was applied in one layer only. The plane was then inclined until the powder slid off as one unit (als Ganzes). Under these conditions, the adhesion and friction measured are those between the powder material and the plane, not those between powder particles. This point was checked by performing the experiment in the manner described by Patat and Schmid. With a single layer of silica sand on a glass plate, the angle of inclination at which individual grains started to slide off was independent of the particle size and smaller than the values of α . With a pile of sand, the bottom layer, remaining after α had been determined, slid off at an angle β given in Table 2. The value of β is essentially independent of the particle size. The spread is comparatively great. As a consequence of the change in the technique, the relation between H and d given by Patat and Schmid is entirely different from that given above, namely $H = kd^{-2}$ for SiC and $H = kd^{-0.7}$ for Al_2O_3 .

The results obtained with Cremer's technique are essentially the same as those discussed in Section 2: adhesion between solid bodies and adhesion between powder particles are effected by water.

4. COALESCENCE OF SOLID BODIES AND PARTICLES

Coalescence of solid bodies and particles has been studied extensively in connection with sintering. Among the numerous papers published on this subject a few that are particularly pertinent to the purpose at hand will be discussed in this section. In addition to those, there are two papers of particular pertinence dealing with crack-healing in NaCl and ice crystals.

Pulliam¹⁸⁾ studied crack-healing in NaCl and MgO crystals by measuring the rate at which a wedge-shaped crack in a single crystal was filled up. Small crystals grew out from the sides of the crack and gradually filled the crack from the root outwards. The phenomenon was attributed to dissolution in moisture followed by precipitation under the influence of capillarity. The same phenomenon was studied by Kliya¹⁹⁾ with ice crystals. The observations and conclusions were the same in both cases. Exploratory experiments conducted in our laboratory with a view to a role of electrostatic charge in the crack-healing mechanism will be discussed in Section 6.

In the experiments just referred to, cracks were produced by cleavage and the healing occurred by precipitation and growth of crystals out from the cleavage plane. This process is easily followed in the microscope at a convenient temperature. Starodubtsev and Timokhina²⁰⁾ studied the same phenomenon by a different technique. NaCl crystals were crushed to 75 to 100 μ , and the grains were sintered by heating for 2 hr at 500°C. The sintered body was then crushed again and examined under the microscope. The grains were held together by bridges grown from the (100) plane (the cleavage plane of NaCl) on each grain in the agglomerate. There were no such bridges observed on the (110) and (111) planes. As an important consequence, it was observed that bridges did not form across the contact surface but between separated surfaces, notably between adjacent (100) faces in the vicinity of the contact. This establishes the crack-healing mechanism as the mechanism of sintering under these conditions.

This effect of crystallographic orientation may explain the rotation of the ice spheres prior to separation reported by Nakaya and Matsumoto, referred to in Section 2. This rotation occurred particularly frequently with ice containing NaCl.

Sintering seems to occur frequently, if not exclusively, by such bridge formation, commonly referred to as necking. The phenomenon has been observed by Chaklader and Roberts²¹⁾ with quartz and by Lee and Parravano²²⁾ with ZnO.

This mechanism is compatible with the essential role of water in solid-state reactions between oxides, silicates, sulfates, etc. Water has been found to be instrumental in these reactions and also in the decomposition of carbonates, sulfates, and other salts. This effect of water is well known in the glass industry. In view of the importance of this effect to the purpose at hand, a few references have been compiled in Table 3.

Table 3. References to observed effect of water in solid state reactions.

<u>Type of reaction</u>	<u>References</u>
Solid-state reactions in general	Eitel ¹⁾ , Taradoire ^{2, 3)}
Glass-making reactions	Eitel ¹⁾ , Ioffe and Shakina ⁴⁾ , Ioffe ⁵⁾ , Raf ⁶⁾ , Ljungquist ⁷⁾
Reactions between oxides	Jander and Stamm ⁸⁾ , Longuet ⁹⁾ , Forestier et al ¹⁰⁻¹³⁾ , Mikhalchenko ¹⁴⁾ , Eubank ¹⁵⁾ , Degueldre ¹⁶⁾ , Arenberg and Jahn ¹⁷⁾ , Borchardt and Thompson ¹⁸⁾
Reactions between oxides and metals	Wickert and Wiehr ¹⁹⁾
Reactions between oxides and carbonates	Jander and Stamm ⁸⁾ , Maekawa and Matsumura ²⁰⁾ , Kröger and Illner ²¹⁾
Reactions between oxides and sulfates	Repa and Canil'chenko ²²⁾
Decomposition of carbonates	Kröger and Fingas ²³⁾ , Preston and Turner ²⁴⁾ , Hüttig and Heinz ²⁵⁾
Decomposition of sulfates	Hüttig and v. Bischoff ²⁶⁾ , v. Bischoff ²⁷⁾ , Briner et al ^{28, 29)} , Pound ³⁰⁾
Reaction of oxide with CO ₂	v. Bischoff ³¹⁾
Reaction of oxide with SO ₂	Postnikov et al ³²⁾

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The conclusions from these observations is almost obvious, namely, the one already mentioned. The reaction consists in the formation of aqueous solution followed by precipitation from the solution. In the case of sintering of MgO or UO₂ (References 15 and 17 of Table 3) as in Pulliam's case of crack-healing in MgO, the process consists in dissolution and capillary precipitation. The reaction of different oxides, e. g., CaO and SiO₂ (Reference 14 of Table 3), consists in the dissolution or hydration of the two oxides to form basic Ca(OH)₂ and acidic H₄SiO₄ and the subsequent capillary precipitation of Ca₂SiO₄. In the thermal decomposition of Na₂CO₃ (References 23 and 24 of Table 3) the reaction consists in the dissolution or hydrolysis of the salt to form NaOH and H₂CO₃ followed by the "precipitation" of CO₂. These reactions were discussed in further detail in a preceding report ²³).

5. FLOW-ABILITY AND AGGLOMERATION IN POWDERS

Agglomeration in powders has been studied extensively in connection with the flow properties of powders. The literature on fertilizers, notably NH₄NO₃, is particularly voluminous. The opinion of the investigators is unanimous: agglomeration and caking are caused by water and can be prevented by drying.

Caking can also be prevented by certain additives in small amounts. These may be of two different types, namely, hydrate formers that take the water away from the powder grains²⁴⁾, and substances that form waterproof coatings on the powder grains²⁵⁾.

Water reduces friction between grains while it increases adhesion between grains. As a consequence, flow-ability of powders may increase with increasing water content to a maximum when the friction decreases more rapidly than the adhesion increases, and then decrease as the adhesion becomes decisive. This effect has been observed by Hofmeister²⁶⁾ in the case of coking coal, which may be expected to show this behavior. The flow-ability had a maximum at 4-5% H₂O and a minimum at 7-10% H₂O. Of course, with large percentages of water, the mixture takes the form of a slurry.

The flow-ability of powders has been studied by measuring the angle of repose. This technique seems to be the one most commonly used. Another technique is the measurement of the force required to lift a disk out of the powder. This technique has been developed and used by Nash et al²⁷⁾.

Craik et al^{28, 29)} measured the angle of repose for starch powders as a function of the amount of MgO added and the humidity of the air. The plot of angle of repose against MgO content shows a minimum at about 1% of MgO. Electron microscope pictures gave evidence to the effect that the starch particles were completely covered by MgO at this composition. The effect of humidity was to increase the angle of repose, particularly when no MgO was added. The addition of MgO conspicuously reduced the humidity effect. Essentially the same results were obtained with sucrose and NaCl powders.

Train³⁰⁾ found that the angle of repose increases with the height of the pile, i. e., with the weight of the powder. This shows that the friction is not negligible at the side of adhesion in these measurements.

Fowler and Wyatt³¹⁾ measured the angle of repose for wheat, rape seeds, sand, polyethylene chips, and basalt chips. The angle of repose increased with the moisture content in the range 0 to 4.5%, except for rape seeds. The deviation was attributed to absorption of moisture as contrasted to adsorption for the other powders.

Nash et al²⁷⁾ have developed a disk-lifting technique that is similar to that of the angle-of-repose technique. A circular disk is immersed in the powder, and the force required to lift it out of the powder is measured. This force gives a measure of flow-ability, but it combines, like the angle of repose, the effects of friction and adhesion. In addition, it depends upon the depth of immersion and thereby upon the weight of the powder above it.

Nash et al measured the force on the disk for a variety of powders at different humidities of the atmosphere and with different deagglomeration agents added. The particle size probably extended over a large range.

Measurements on Carbowax 6000 and Carbowax + Tri-calcium Phosphate at 5, 59, and 80% relative humidity showed that the force decreased with increasing humidity. Measurements on saccharin, Carbowax 6000, and Span 60 in vacuo and in the ambient showed that the force was greater in the ambient than in vacuo for the first two materials, whereas the reverse was found with Span 60. The difference was attributed to sublimation of Span 60 and the formation of new surfaces in the process. The reduction in the force on evacuation was attributed to removal of water.

Nash et al also studied the effect of additives upon the angle of repose. Since it was associated with the electrostatic charge on the powder particles, these measurements will be discussed in Section 6.

Although the angle-of-repose and the disk-lifting techniques do not separate friction and adhesion, the results obtained with these techniques are well compatible with those of Cremer and of our laboratory. They show, in addition, that the two effects are of comparable magnitudes, and that a change in the experimental conditions may change the flow-ability in either direction, depending upon its relative effects upon friction and adhesion.

6. EFFECT OF ELECTROSTATIC CHARGE

An effect of electrostatic charge upon agglomeration and adhesion is strikingly absent in the evidence presented in the preceding sections. In the experiments of Stone, Tomlinson, Bradley, and Harper, discussed in Section 2, an effect of electrostatic charge upon the adhesion between glass or quartz spheres was carefully looked for but not found. In view of the ease with which the electrostatic attraction is shown to exist, e. g., by means of a fountain pen and a small piece of paper, this absence of an effect of electrostatic charge upon agglomeration and adhesion may appear puzzling. Exploratory experiments conducted in our laboratory seem to indicate an explanation.

NaCl crystals were cleaved to pieces of about 4 mm edge. In this process the pieces acquired charges. Two pieces were brought close to each other on a microscope slide at an angle that would simulate a wide crack, say, 20° . Distilled water was introduced in the "crack" by means of a No. 30 hypodermic needle. When the crystals were charged, the water drop refused to go into the crack but jumped to one or the other of the two crystals. After, as a rule, three drops, enough water had been added for flow into the crack to occur, but most of the water seemed to go along the sides of the crystals onto the slide and underneath the crystals. At this stage the crystals were virtually floating on a film of water. The interesting phenomenon was then observed that the two crystals snapped together and closed the gap between them. This occurred very rarely when the distilled water was replaced by a saturated NaCl solution and then only when the two crystals were comparatively close together from the start. When this phenomenon did not occur, i. e., when the initial distance was too large for attraction or when NaCl solution was added, the interspace filled up by precipitation and crystal growth as reported by Pulliam. This process occurred more rapidly with NaCl solution than with distilled water and was in the latter case preceded by an induction period, in which the formation of NaCl solution presumably took place.

An interesting observation made in these studies is that crystals do not always grow from the two surfaces in the same abundance and at the same rate. After addition of distilled water, the crystal growth may occur persistently predominantly from one surface; after addition of NaCl solution, the crystal growth may occur from one surface in a short initial period but then become equal on the two surfaces.

It may be added to these observations as an explanatory note that a water drop released from a hypodermic needle held above a charged insulator takes on a charge that is proportional to that of the insulator but of opposite polarity. Thus, if the two NaCl crystals have the positive charges q_1 and q_2 , the water drop takes on a charge $-k(q_1 + q_2)$, the proportionality constant k depending upon the distance between needle and crystal and on other parameters. The value of k is the same for distilled water and aqueous solutions. These phenomena were studied in a recent investigation in this laboratory³²).

When the water drop falls off the needle, it is attracted by the two crystals by forces proportional to $q_1(q_1 + q_2)$ and $q_2(q_1 + q_2)$, respectively. It lands on the crystal with the higher charge, say, q_1 . The two charges q_1 and $-k(q_1 + q_2)$ partly neutralize each other, and

the new charge is $q_1 - k(q_1 + q_2)$. Accordingly, one of the two crystals will change the polarity of its charge after receiving one or several drops, so that the two crystals become oppositely charged. This requires, as a rule, three drops of water in this particular experiment.

There is a simple explanation to the observations just reported. The NaCl solution being conductive, the electrostatic charges on the crystal surfaces neutralize each other by a current through the solution. There can, therefore, be no large electrostatic force when NaCl solution is added. The distilled water is a comparatively poor conductor and does not cause neutralization of the charges. But after some time the distilled water has dissolved enough NaCl to cause a discharge. Thus, whether distilled water or NaCl solution is added makes no difference to the ultimate result; the electrostatic attraction disappears. It is a matter of course that the discharge occurs more rapidly at a short distance, i. e., when the two crystals are in contact with each other, than when they are a distance apart.

It follows that electrostatic attraction is a long-range force only, and that it does not contribute to the adhesion between bodies or particles in contact.

It was found in a previous investigation³³⁾ that electrostatic charges had a conspicuous effect upon the settling of aerosol particles. Aerosol particles settling on a millipore filter agglomerated in patterns corresponding to the charge distribution on the filter surface. Aerosol particles settling on a glass slide formed chain structures with alternating positive and negative particles in the chain. However, there were no such chains when the aerosol particles settled on a conductive surface, chromium foil, or aluminized glass. It was also established by the use of a vaseline coating on the glass slide that the particles did not form the chain structures while airborne but did so after deposition, if they were free to move over an insulating surface. It follows from the comparison between insulating and conducting surfaces that the charged particles transfer their charges fairly readily to a conducting surface.

It seems safe to conclude from this discussion that there is no effect of electrostatic force upon adhesion and agglomeration in moist soluble salt powders. The force would be operative and would persist only if the powder is non-conductive. However, perfect insulators do not seem to acquire charge in friction.

It has been found by several investigators³⁴⁻³⁶⁾ that perfectly insulating liquids do not acquire electrostatic charge when a gas is bubbled through them. At very low conductivities the charge is proportional to the conductivity. This should apply to triboelectrification in general, because the charge produced in friction must necessarily spread over an area, and not be confined to a point of contact, in order to reach an appreciable magnitude. Accordingly, perfect insulators do not acquire charge in friction, and particles of perfect insulators would, therefore, not be charged under ordinary conditions.

It was shown in a recent investigation³⁷⁾ that the coalescence of two liquid drops in contact occurs at a rate that is proportional to the voltage between the two drops or, at high voltages, to its square. This effect of electrostatic charge is not related to the electrostatic force but rather to the field or, at high voltages, to the current. It seems possible that similar effects may occur with solid particles. So far, little evidence has been gathered to the existence of such effects with solid particles, but the observations made with the NaCl crystals encourage investigations in this area.

Nash et al³⁸⁾ have studied the effect of electrostatic charge upon the flow-ability of powders. The measured quantity was the angle of repose, i. e., the angle formed when the powder flows into a pile. It was found that Carbowax 6000 had an angle of repose of 58° , whereas the same powder after addition of 1% of Cab - O - Sil as a deagglomeration agent had an angle of repose of 44° . Observations were reported, which indicate that the effect of the Cab - O - Sil was related to the electrostatic charge on the powder. The effect of Cab - O - Sil on saccharin powder was much less, namely, to reduce the angle of repose from 58° to 56° . A number of other deagglomeration agents were tested with saccharin. Some of them gave a decrease, others gave an increase in the angle of repose. Shear strength measurements showed similar effects, but the order of effectiveness was different in the two cases. Furthermore, certain additives gave a reduction in shear strength and an increase in angle of repose or vice versa. As pointed out in Section 3, these measurements do not separate the effects of friction and adhesion. The results indicate that different deagglomeration agents have different relative effects upon friction and adhesion.

The evidence available is not conclusive, but it is compatible with a primary effect of water and a secondary effect of electrostatic field or electric current in agglomeration and adhesion.

7. APPLICATIONS TO AEROSOLS

The evidence considered in the preceding sections is directly applicable to aerosol particles as to the nature of the bonds between particles adhering to each other and as to the mechanisms by which these bonds are formed. In the case of the crack-healing mechanism, rate processes are involved, namely, dissolution and precipitation. The rates of these processes depend upon the rate of dissolution of the particles, the distance between suitable crystallographic faces at contact, the amount of water present, and, maybe, the electrostatic charge. These factors are hard to simulate in model experiments, and it is correspondingly hard to estimate the importance of this mechanism relative to that of the other mechanisms of adhesion. It appears reasonable to assume, however, that adhesion by other means than coalescence (crack-healing), cannot be strong and permanent to the extent of significance to aerosol stability.

If it is thus assumed that coalescence is the sole contributor to agglomeration of solid aerosol particles, agglomeration requires the coalescence to be rapid enough to occur in the period of contact between particles in collision. This introduces another rate element, namely, the time of contact. One may then assume that an elastic collision between two particles has too short a time of contact, and that coalescence therefore requires temporary bonds to be formed and to prolong the contact enough for coalescence to occur.

It appears that electrostatic charge may well be decisive to agglomeration under these conditions. In the case of low conductivity across the contact, the charge leaks off slowly, and the electrostatic force may persist for an appreciable length of time. Furthermore, the electrolytic effect of the discharge current may accelerate the coalescence. No direct evidence on these issues could be found in the literature. The reasoning is therefore rather speculative.

8. CONCLUSION

The experimental evidence on the adhesion between solid bodies and between powder particles is conclusive to the extent that water is the cause of adhesion under normal conditions. It strongly suggests three different mechanisms for adhesion by water:

- a. Capillary forces (surface tension) in the case of comparatively thick films of water on insoluble solids.

- b. Intermolecular forces (hydrogen bonds) between water molecules and OH groups in the solids in the case of comparatively thin (monomolecular) films on insoluble solids.
- c. Coalescence by dissolution and precipitation in the cases of thin or thick films of water on soluble solids.

There is no or negligible contribution to adhesion by electrostatic forces per se, except for a short period of time after contact, but electrostatic charge may affect the adhesion by water through the field or through the discharge current in charge neutralization. The evidence on the role of electrostatic charge is not conclusive, however, as to existence and magnitude of these effects. Further experimental work in this area is required.

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14. KEYWORDS <table border="0"><tr><td>Solid aerosol particles</td><td>Water</td><td>Capillary forces</td></tr><tr><td>Intermolecular forces</td><td>Coalescence</td><td>Electrostatic charge</td></tr><tr><td>Adhesion between solid bodies</td><td>Adhesion between powder particles</td><td></td></tr></table>				Solid aerosol particles	Water	Capillary forces	Intermolecular forces	Coalescence	Electrostatic charge	Adhesion between solid bodies	Adhesion between powder particles	
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